Thanks for pointing out these issues. The regressions could certainly be carried out by season or month, and have a clear advantage of doing so if it reduces the noise in the EC-τ_R relationship. This is, however, not the case for our dataset. The seasonal segregation does not improve the correlation or scatter around the best-fit lines, as shown in the supplemental figure (Fig. S-2), suggesting that daily variability (resulting from changes in chemical composition and/or measurement uncertainty) is comparable to the seasonal variability in the EC–τ_R relationship. Segregating data by month provides no clear improvement, either. It is reasonably representative to report one regression and statistical test that consider year-round data, providing that the data are uniformly distributed throughout a year. This also corresponds to our trend analysis technique based on annual data.

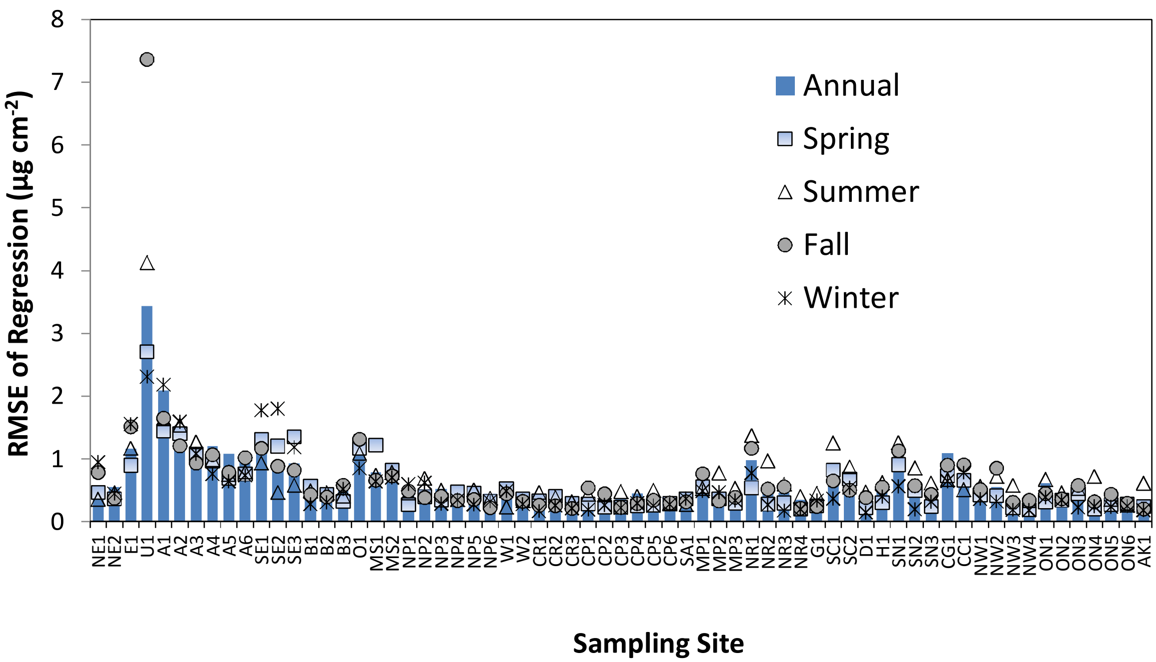

Figure S-2. Scatter around the best-fit line (Eq. [5] in the manuscript), by site and by season, compared to that for site-specific annual (year-round) data. Scatter is evaluated through the root-mean-square-error (RMSE) between the measured and predicted dependent variable (i.e., EC). See Table S-1 for definitions of site code.

In the revised manuscript, we added a new paragraph (Lines 224–228) to explain the situation:

> (Line 224-228) "The regression analysis was also carried out by season. However, such seasonal segregation does not reduce scatter around the best-fit lines (Figure S-1, Supplement). This suggests daily variability (due to changes in chemical composition and/or measurement uncertainty) comparable to seasonal variability in the EC-τ_R relationship and that year-round regression analyses are reasonably representative of all cases."

As to the changes in the optical monitoring system and definition of EC, BC, and LAC, this is addressed in the responses to the comments from Reviewers #1 and #2. As shown in the Response to Reviewer #1, the principle and geometry of reflectance measurement remains the same, i.e., laser beam is directed to the sample through a coaxial optical fiber and a quartz light pipe (perpendicular to and ~ 2 mm from the sample) by which the reflected light is acquired. We have explained that the instrumental upgrade likely influences EC measurement more than τ_R measurement. Lines 58-61 of the Introduction section have been revised to clarify the definition of EC and BC:

> (Line 58-61) "Elemental carbon (EC), a light-absorbing carbon (LAC) component as determined by thermal/optical methods, is the dominant aerosol fraction that absorbs visible radiation in the troposphere (Andreae and Gelencsér, 2006). This fraction is often termed "black carbon" (BC) if quantified by optical or photoacoustic methods (Moosmüller et al., 2009)"

References

Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131-3148, 2006.

Moosmüller, H., Chakrabarty, R. K., and Arnott, W. P.: Aerosol light absorption and its measurement: A review, J. Quant. Spectrosc. Radiat. Transfer, 110, 844-878, 2009.